



The HVEE Tandetron AMS system at Nagoya University

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Abstract

In 1996/97, we installed a second-generation AMS system (model 4130-AMS) manufactured by HVEE, B.V., The Netherlands. Although we encountered a lot of problems during the installation and tuning up of the machine, our new system has now reached good stability and reproducibility in performing $^{14}\text{C}/^{12}\text{C}$ and $^{13}\text{C}/^{12}\text{C}$ measurements. According to the reproducibility tests conducted in January 1999, where six graphite targets prepared from HOxII standard material were measured, the standard deviation for $^{14}\text{C}/^{12}\text{C}$ reproducibility is around 1.6‰ (comparable with counting statistics) and that for $^{13}\text{C}/^{12}\text{C}$ is 0.28‰. The background level of the AMS system was measured using a pure graphite (commercial graphite rod). It lies between 50 and 55 ka BP. We are still on the way to reduce the background. However, we are now prepared to start routine measurements of unknown-age samples. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Since 1983, a Tandetron manufactured by General Ionex Corporation (GIC), USA, is in operation at the Dating and Material Research Center at the Nagoya University [1]. Being the heart of our old accelerator mass spectrometer system, it has been used to measure about 8500 ^{14}C samples from various research fields such as cultural properties, archeology, anthropology, geog-

raphy, geology, hydrology, oceanography, limnology and glaciology. It was also used to measure ^{14}C concentrations of environmental samples related to atmospheric science, wood science and soil science [2–5].

In 1996/97, we have installed a second radio-carbon dating system. It is a new second-generation AMS system around a Tandetron (Model 4130-AMS) built by High Voltage Engineering Europe, B.V., The Netherlands [6,7]. Two similar HVEE AMS systems have been installed successfully at the University of Groningen, The Netherlands, and at the Christian-Albrechts-University at Kiel, Germany [8,9]. They have already proven excellent performance in ^{14}C -AMS measurements giving $^{13}\text{C}/^{12}\text{C}$ ratio with a reproducibility better

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than 0.1‰. The statistical uncertainty and the reproducibility of the $^{14}\text{C}/^{12}\text{C}$ ratios measured were 1.5–2.2‰ and 3.0‰, respectively. These results implied that the error of the ^{14}C ages has been reduced to about ± 25 yr. In Asia, another two sets of HVEE Tandetron AMS systems have been installed: one at the Mutsu Branch of Japan Atomic Energy Research Institution in Japan (1997), and another at the Seoul National University in Korea (1998).

We describe here the performance tests carried out in our system and some results from a first application: the ^{14}C dating of geological samples.

2. System description

The main improvements in the HVEE AMS system compared with the old GIC system [1] are:

1. A high-intensity cesium sputter ion source (Model 864B) is provided with the new system, so that the ^{14}C counting rate in the rare isotope detector is almost an order of magnitude higher than the rate gained with the old source (HICONEX 844). In addition, since up to 59 targets (samples) can be loaded at the same time and sample by sample is measured automatically under computer control, measurements are conducted more efficiently.
2. The carbon isotopes $^{12}\text{C}^-$, $^{13}\text{C}^-$ and $^{14}\text{C}^-$ are injected into the accelerator simultaneously using a recombinator system achieving high accuracy measurements.
3. ^{14}C -AMS can be performed at a terminal voltage of 2.5 MV, which gives the maximum yield for stripping C^- to C^{3+} . The terminal voltage is stabilized using a slit-feedback system monitoring the energy-dependent position of the $^{13}\text{C}^{3+}$ ions after the high-energy analyzing magnet. The reached stability of the terminal voltage $\Delta V/V \approx 6 \times 10^{-4}$ [6] is furnishing highly stable isotope-ratio measurements.
4. In order to separate the $^{14}\text{C}^{3+}$ ions from the various background ions, the new rare isotope detector (ionization detector) measures the total kinetic energy, E_{tot} , and the residual energy, E_{res} , of the ions (isobutane, ca. 15 mbar). Rejecting the background ions very efficiently,

the new system enables us to measure ^{14}C ages as old as 60 ka BP.

As a result of these improvements, the achievements with the new system are:

1. The measurement error of the ^{14}C age may be reduced to ± 25 yr with data collecting times of a few tens of minutes for a modern carbon sample of 1 mg.
2. The measurement can be performed fully automated.
3. More than 3000 samples can be measured a year, provided that samples can be prepared in time.

3. Performance tests

3.1. Reproducibility of $^{14}\text{C}/^{12}\text{C}$ and $^{13}\text{C}/^{12}\text{C}$ ratio measurements

During the installation of the machine, we have experienced many troubles. One of the most serious problems was that the ion beam leaving the accelerator was too low. The steerers at the exit of the machine were not strong enough to bend the beam back to its right position. We sent the accelerator tubes back to HVEE where they were carefully scrutinized. We also replaced the steerer power supply by a stronger one. Having tackled all the problems, we were able to run the performance tests for the new spectrometer in January 1999. $^{14}\text{C}/^{12}\text{C}$ and $^{13}\text{C}/^{12}\text{C}$ ratios were measured on two consecutive days. We used six graphite targets prepared from the HOxII oxalic acid standard material by the AMS group of the Christian-Albrechts-University, Germany [9]. Each target was measured five times the first day and nine times the second day. The duration of one measurement was 10 min. At a terminal voltage of 2.5 MV, the ^{14}C count rate was typically 80 cps. The $^{12}\text{C}^{3+}$ and $^{13}\text{C}^{3+}$ currents were 280 and 290 nA, respectively. The results of these measurements are listed in Table 1. The standard deviation (1σ) of the $^{14}\text{C}/^{12}\text{C}$ ratios of the six targets measured the first day was 1.7‰ (better than the uncertainty of 2.1‰ due to counting statistics). The standard deviation of the corresponding $^{13}\text{C}/^{12}\text{C}$ ratios was 0.33‰ (Fig. 1). On the second

Table 1

Reproducibility of $^{14}\text{C}/^{12}\text{C}$ and $^{13}\text{C}/^{12}\text{C}$ ratio measurements with the new HVEE Tandetron AMS system at Nagoya University

Sample #	^{12}C current (nA)	^{13}C current (nA)	^{14}C (counts)	$^{13}\text{C}/^{12}\text{C}$ ($\times 10^{-2}$)	$^{14}\text{C}/^{12}\text{C}$ ($\times 10^{-12}$)
<i>Acceptance test (first day)^a</i>					
A01	284.3	296.0	219 550	1.15704	1.52050
A02	282.6	294.3	217 806	1.15712	1.51742
A03	280.6	292.0	215 793	1.15646	1.51440
A04	291.8	303.9	224 229	1.15728	1.51368
A05	281.1	292.9	216 300	1.15760	1.51456
A06	276.3	287.7	212 897	1.15690	1.51732
Average			217 763	1.15707	1.51631
Statistical error (1σ)			0.00214		
Relative standard deviation (1σ)				0.00033	0.00171
Error of ^{14}C age (1σ) (yr)			17.2		13.2
<i>Acceptance test (second day)^b</i>					
B01	289.0	300.9	399 709	1.15673	1.51316
B02	282.9	294.7	391 527	1.15719	1.51378
B03	287.4	299.4	399 039	1.15744	1.51908
B04	279.4	290.9	387 748	1.15696	1.51827
B05	280.5	292.0	388 472	1.15658	1.51491
B06	284.1	295.9	394 038	1.15716	1.51710
Average			393 422	1.15701	1.51605
Statistical error (1σ)			0.00159		
Relative standard deviation (1σ)				0.00028	0.00162
Error of ^{14}C age (1σ) (yr)			12.8		13.0
<i>Summary of the test runs conducted from 23 to 27 July 1999^c</i>					
C01	108.8	113.4	183 902	1.1582	1.5105
C02	109.6	114.3	185 929	1.1581	1.5177
C03	109.2	113.9	185 479	1.1582	1.5197
C04	109.6	114.3	185 971	1.1584	1.5185
C05	111.1	115.8	188 593	1.1579	1.5195
C06	111.7	116.5	189 692	1.1582	1.5198
Average			186 594	1.1581	1.5176
Statistical error (1σ)			0.00232		
Relative standard deviation (1σ)				0.00013	0.00233
Error of ^{14}C age (1σ) (yr)			18.6		18.7

^a Five cycles of 10-min runs.^b Nine cycles of 10-min runs.^c Eleven cycles of 10-min runs.

day the standard deviations were 1.6‰ (1.6‰) and 0.28‰, respectively. The acceptance tests demonstrated that our new AMS system can perform high-precision measurements of $^{14}\text{C}/^{12}\text{C}$ and $^{13}\text{C}/^{12}\text{C}$ ratios.

Just after the acceptance tests, our Tandetron suffered from troubles affecting the high-voltage generating system. The high voltage was not stable and the machine was not able to reach 2.5 MV.

The reason for this was found in the diode stack generating the high voltage. About one-fourth of the 1422 diodes and one-tenth of the 158 resistors were defective. After the replacement of the damaged parts, we tested the performance of the facility anew. However, we limited the beam current to one-third of that used for the acceptance tests. The results of these measurements proved that the stability and the reproducibility were again excel-

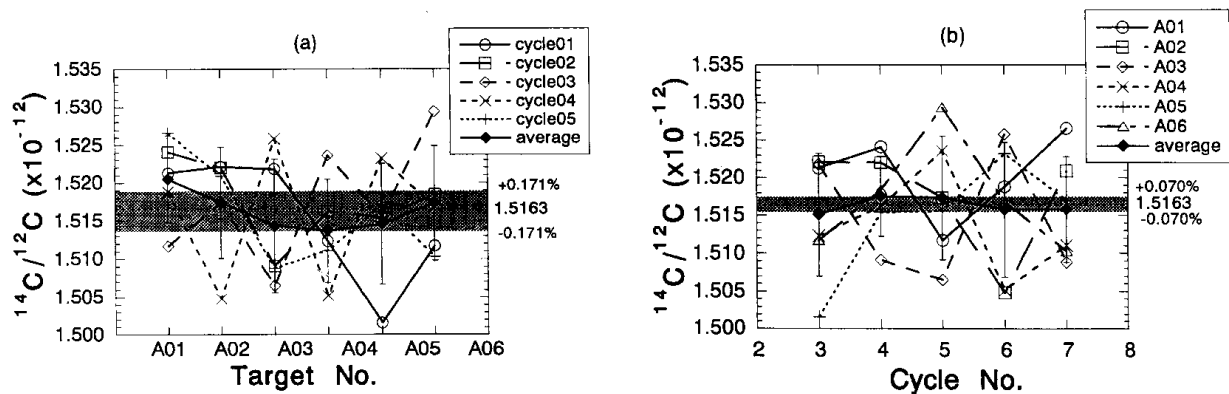


Fig. 1. Reproducibility of the $^{14}\text{C}/^{12}\text{C}$ ratio measured for HOxII standard material put into six target holders (first day of acceptance tests, January 1999): (a) the $^{14}\text{C}/^{12}\text{C}$ ratio measured during five cycles together with the corresponding mean value of the individual targets plotted versus target number; (b) the $^{14}\text{C}/^{12}\text{C}$ ratio of six different targets together with the corresponding mean values of the six targets plotted versus cycle number. Cycle numbers 1 and 2 are cleaning runs and are excluded for the analysis. The shaded band depicts the mean value (straight dashed line) of all the targets and the relative standard deviation.

lent (Table 1). We reached similar results as during the acceptance tests.

3.2. ^{14}C background of the AMS system

The ^{14}C background level was estimated using the IAEA standard material C-1 and a commercial graphite rod (see Table 2). In an evacuated bottle, the C-1 carbonate (Carrara marble) was reacted to CO_2 by adding H_3PO_4 . The CO_2 was then purified and converted to graphite using a routine procedure [2,3]. The rod graphite gave the machine background of 50–55 ka BP, while the graphite from the IAEA C-1 standard supplied information on the ^{14}C contamination due to the sample preparation procedure for carbonate (46–49 ka BP).

4. Application test

4.1. ^{14}C dating of bottom sediments from Lake Baikal

As an application test, we have measured ^{14}C ages of the total organic carbon extracted from samples of a sediment core. From January to April 1998, the core (BDP98) was drilled to a depth of 600 m at the Academician Ridge (53°44'40" N, 108°24'30" E) of Lake Baikal, Siberia, Russia. The sediments started 325 m below the lake's surface. The sediment samples used for dating were carefully separated from the inner part of the core and wrapped in aluminum foil before putting them into individual plastic bags. At the laboratory in Russia, they were cooled and kept frozen in a

Table 2

^{14}C background of the new HVEE Tandem AMS system at Nagoya University, and that of the sample preparation procedure, estimated by using commercial rod graphite and IAEA C-1 standard material (carbonate, Carrara marble), respectively

Sample #	Sample material	^{14}C conc. (pMC, %)	Apparent ^{14}C age (yr BP)
1	Rod-G-1	0.16±0.03	51 750±1320
2	Rod-G-2	0.20±0.03	49 860±1230
3	Rod-G-3	0.15±0.02	52 510± 930
4	Rod-G-4	0.21±0.04	49 720±1440
5	Rod-G-5	0.11±0.03	55 030±2420
6	IAEA C-1-1	0.23±0.04	48 750±1240
7	IAEA C-1-2	0.33±0.05	46 020±1320

refrigerator. The frozen samples were then shipped to our AMS facility.

At our laboratory, the sample material was pretreated with 1.2 M HCl at 80°C for 2 h and then rinsed with distilled water to completely remove any acid fraction. This treatment was performed twice to get rid of the carbonates. The sample was then dried at 90°C. After this pretreatment the remaining material, containing about 2 mg carbon, together with an appropriate amount of CuO, was filled into a Vycor tube. After evacuating, the tube was flame sealed and heated to 900°C for 2 h combusting the sample material. The resulting CO₂ was purified in a glass vacuum line using liq. N₂, C₂H₅OH–liq. N₂ mixture (–100°C) and N-pentane (–130°C). The purified CO₂ was then converted to graphite by reducing it catalytically on Fe powder. For this purpose, the CO₂ was sealed together with H₂ in a Vycor tube and kept at 650°C for 6 h [10].

The ¹⁴C/¹²C and ¹³C/¹²C ratios of the sediment samples together with a HOxII standard were measured using the new AMS system. Each sample was measured for about 10 min. The typical uncertainty due to counting statistics was 0.7% (HOxII). In order to correct the ¹⁴C/¹²C ratios for isotopic fractionation, the δ¹³C values of the individual samples were measured by a conventional multi-collector mass spectrometer (Finnigan MAT252).

The results are shown in Fig. 2. Only the upper part (from the sediment surface down to 370 cm)

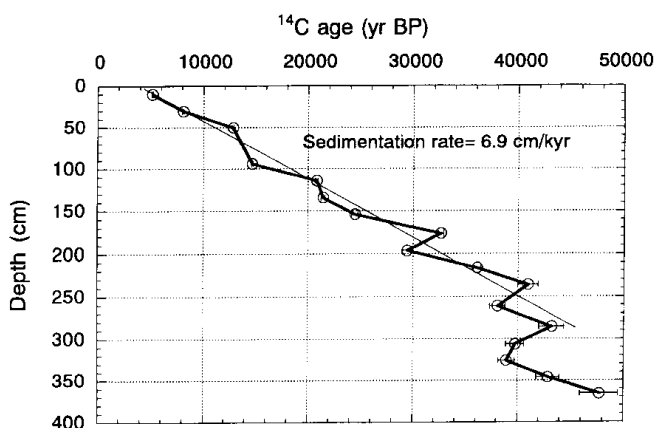


Fig. 2. ¹⁴C ages versus depth for the DBP98 hole 1 core sediment. The total organic carbon separated from the sediment was used for dating (see text).

of the 600 m core has been dated. The ¹⁴C age increases linearly with depth (as far as 230 cm), but looks saturated around 40 ka BP. We suspect that this may be due to the carbon contamination during the sample preparation procedure. The calculated mean sedimentation rate for the first 230 cm of the core is 6.9 cm/ka. The reliability of the measured ¹⁴C ages will be discussed elsewhere together with paleo-environmental changes that are studied and revealed by investigating the sediment from the same core.

4.2. ¹³C/¹²C ratio measured with the new AMS system

The ¹³C/¹²C ratios of the Lake Baikal sediment samples were measured using the HVEE AMS system and normalized by the also measured ¹³C/¹²C of the HOxII standard material. The results were compared with the corresponding ¹³C/¹²C ratios got with the Finnigan MAT252 mass spectrometer (Fig. 3). The correlation is not perfect, however, the ¹³C/¹²C ratios measured with the HVEE machine do not differ much from the MAT252 values. So they can potentially be used for the correction of the ¹⁴C ages. We should continue to try to minimize the difference between ¹³C/¹²C measured by these two different instruments.

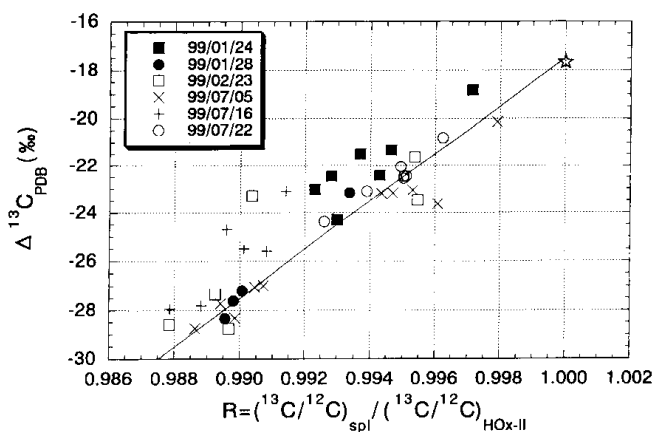


Fig. 3. Comparison of ¹³C/¹²C values measured with the new AMS system and with a Finnigan MAT252 stable isotope mass spectrometer. The star indicates the value of the HOxII standard material having a δ¹³C of –17.5‰.

5. Concluding remarks

Although we have encountered a lot of unexpected problems, the new AMS system has proven to provide us quite high-quality ^{14}C data, and we will be able to start routine measurements soon. We have started further reliability tests with the HVEE Tandatron using IAEA standard material and some other samples of known age. Our initial application programs of the high-performance AMS system are:

1. annual ring samples of trees related to volcanic eruptions are to be dated precisely and with high resolution. The ^{14}C ages will be used for an accurate age determination of the volcanic eruptions by the wiggle-matching method;
2. precise ^{14}C dating of historic cultural properties and comparison of calibrated ^{14}C ages with the historical ages of the investigated properties [11,12];
3. precise measurement of atmospheric tracer as CO_2 , CH_4 and CO , in order to analyze the carbon cycle in our environment.

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